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PRELIMINARY COMMENT

The attached manuscript is a first draft of a paper that will be submitted for publication. This represents the main emphasis of the work performed on this Contract during the past year.

The Hollow Cathode Helium-Fluorine Laser*

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ABSTRACT

It is possible to obtain uniform stable, long pulse excitation ($\geq$ 100 μsec) in gas mixtures involving highly electronegative constituents (SF_6, CCl_4, NF_3, I_2). Such a system was used to investigate the atomic Fluorine laser. In the hollow cathode, lasing on Fluorine transitions in the doublet system lasted for up to 80 μs with no signs of the self-termination as reported previously in positive column devices. The excitation process of the laser appears to depend heavily upon the Fluorine donor utilized. For instance, a single step process is involved when NF_3 is used whereas a two step process is evident for SF_6. The details will be discussed.

I. Introduction

The hollow cathode glow discharge exhibits certain characteristics not found in a normal, positive column discharge. Certain of these properties would appear to be eminently advantageous to the excitation of various gaseous species for the purpose of producing a laser. In this work, the properties of the hollow cathode discharge were exploited to excite some particularly difficult gas mixtures (involving highly electronegative gases), and to pump a laser in atomic Fluorine.

II. Excitation of Electronegative Gas Mixtures

The feature of the hollow cathode discharge which makes it highly useful in exciting mixtures of gases containing electronegative species is the highly non-Maxwellian nature of the electron energy distribution.[1-3] Borodin and Kagan[4] in comparing the electron energies in a hollow cathode discharge in He to a similar discharge excited in a positive column found a greater number of high energy (> 19.8 eV) electrons in the hollow cathode. Furthermore, Belal and Dunn[5] found a significantly higher proportion of Helium ions than metastables in a Helium hollow cathode discharge. This non-Maxwellian electron energy distribution is the result of high energy, "beam," electrons from the cathode fall region which cause ionization and excitation in the negative glow. The beam excitation in a hollow cathode makes this device analogous to an electron-beam controlled device in which virtually any type of gaseous medium (including highly electronegative gas mixtures)

can be excited. In Fig. 1 a hollow cathode device is shown in which a uniform, stable glow discharge was obtainable in a variety of electronegative gases (NF_3, SF_6, CCl_4, I_2). This device consists of two concentric, cylindrical electrodes with the inner cathode slotted to allow a current path from inside the cathode tube to the outer anode similar to that used by Schuebel. The cathode is a 1 cm. dia. x 50 cm. stainless steel tube and the anode is 1.25 cm. di . x 50 cm. stainless steel. In the hollow cathode mode, the negative glow region fills nearly the entire inner volume of the cathode. The photograph in Fig. 1 shows the voltage and current pulses for a discharge in pure SF_6. As indicated by the photograph there is no tendency for the discharge to evolve into a high current constrictive mode as would be the case in a positive column discharge in SF_6. This ability to obtain stable, long pulse discharges in highly electronegative gas mixtures makes this device a logical medium for investigating any laser using the halide gases.

III. The Atomic Fluorine Laser

Of the atomic halide lasers, the Fluorine laser appears to be most interesting with its multitude of lines in the red and near infrared. Several authors[7-15] have previously reported work on the Fluorine laser - all in positive column devices. Lasing has been observed on thirteen lines in the doublet and quartet systems of Fluorine ranging from 6239 Å to 7802 Å. These lasers (ranging from low pressure long bore tubes[7,8,12] to high pressures, fast, TEA devices[10,13-15]) have been operated in mixtures of He plus some Fluorine donor (e.g. SF_6, NF_3, CF_4, HF, F_2). Helium pressure ranges from less than one torr to greater than an atmosphere in the case of the fast TEA configuration, with Fluorine donor

concentrations of 50μ to 8 torr. Lasing on the quartet lines has only been observed in the high pressure devices, while the doublet system lases in both the high and low pressure devices.

Figure 2 shows an abbreviated energy level diagram of the doublet system in atomic Fluorine along with the dissociation limit of SF_6 and the $He(2^3S_1)$ metastable. Pumping of the upper laser states in Fluorine is thought to occur according to the following dissociative excitation process:[11]

$$He(2^3S_1) + SF_6 \rightarrow F^*(3_p 2P) + He(1'S) + SF_5^*. \qquad (1)$$

This reaction has a rather large defect ($\sim$ 2 eV), but is thought likely to occur for the following reasons:[16] (1) due to the complexity of the SF_6 molecule, there are many degrees of freedom available to absorb the excess energy; and (2) the probability for the reverse reaction involving the polyatomic fragment SF_5, is very low.

Although SF_6 is mentioned in the previous discussion, other Fluorine compounds appear to be more suitable donors. In particular, NF_3, HF, and F_2 have produced better results than SF_6.[8,10,12,13] In the case of HF, its 5.85 eV dissociation limit places the upper Fluorine excited levels within 0.1 eV of the 2^1S_o metastable of He.[8]

In spite of the encouraging amount of work previously reported on the Fluorine laser, several severe limitations have hindered the usefulness of this device. A major encumbrance to this device is its short pulse limitation. According to Kovacs and Ultee,[7] bottlenecking occurs in the lower state due to trapping on the 955 and 956 Å resonant transitions. This limits operation of the device to 1-2 μs. Jeffers and Wiswall[8] reported a solution to this problem by using HF as the

Fluorine donor since, with free Hydrogen available in the discharge the following reaction can occur:

$$F(3s^2P) + H(n=1) \rightarrow F(2p^2P_o) + H(n=5) \qquad (2)$$

which leads to an effective lower state lifetime of 10^{-7} - 10^{-8} seconds (including the effects of resonant trapping). Nonetheless, their laser operated in the afterglow of the excitation pulse, thus foregoing any CW operation. In the experiment reported below, lasing for upwards of 80 μsec have been obtained.

Description of Experiment

The experimental setup itself is quite straightforward. The discharge tube is designed so that the laser gas mixture can be flowed through it in a continuous manner using a small forepump. In this way the gas mixture is constantly replenished, although the flow rate is slow enough (< 25 ℓ/min) that the particular volume of gas is static for the duration of a single pulse (t < 1 msec). The gases are pre-mixed in a small manifold with an oil manometer to measure gross pressures.

The gas mixture itself varies over a considerable range of pressure which depends on the dimensions of the particular hollow cathode tube being used. For the smaller diameter (0.9 cm) cathode, the He pressure is about 10 torr with 100-200 μ of the Fluorine donor. A larger diameter cathode (2.3 cm) is operated with about 3 torr He and 70-150 μ of the Fluorine donor. In this work SF_6 is the most commonly used donor because of its availability and ease of handling. NF_3 which has proven to be a better donor is used whenever available.

The electrical excitation for the hollow cathode discharge is provided by two different types of pulsing circuits. In one, a hydrogen thyratron is used in conjunction with a transmission line to deliver a fixed length pulse (< 5 μs) of up to 300 amps. The second circuit is a commercial Velonex pulser which provides a rectangular and continuously variable width pulse with a maximum current capability of about 15 amps. Detection and measurement of the laser signal and spontaneous emission are accomplished using either a PIN5 photodiode or a GaAs photomultiplier in conjunction with a monochromator.

<u>Long Pulse Operation</u>

A most interesting feature of the hollow cathode Fluorine laser is its capability to run in a long pulse mode at medium power levels (1-10 watts), along with the potential to extend this to CW operation. Figure 3 shows a photograph of the laser pulse along with the discharge current pulse in which lasing occurs for about 80 μs. The fact that lasing lasts for such a long period of time implies that there are no fatal limitations such as bottlenecking on the lower state. The question, then, is what are the primary physical processes which prevent the ultimate limitation to long operation of the Fluorine laser?

The data given below suggests that the depletion of the Fluorine donor is a major cause of termination of laser action.

Figure 4 shows the spontaneous emission on the 7128 Å transition for three different values of discharge power. As can be seen by these curves, the rate of decay of the spontaneous emission is obviously a function of input power. For the lowest value of excitation, the spontaneous emission closely follows the input power while for increasing values of excitation, the decay rate of the fluorescence becomes more rapid.

While trapping could have a very serious effect on the laser performance, one would be hard pressed to assign that as the cause of the decrease of spontaneous emission from the upper laser level. There is no trapping on the 7129 Å transition - quite to the contrary - there is gain (although it is miniscule across the diameter of the tube). There is no way that trapping on the 956 Å transition can affect the spontaneous emission at 7128 Å when the population is inverted. Indeed, the time dependence of this emission suggests a depletion of one of the constituents involved in the excitation of the upper state.

For instance, the excitation F^* with NF_3 as the donor (corresponding to the data of Fig. 4) is thought to proceed by the following route:

$$He^* + NF_3 \rightarrow F^*(3p^2P) + He(1'S) + NF_2 \qquad (3)$$

with He^* being one of the metastable states. The spontaneous light presented in Fig. 4 is, of course, a relative measure of F^* which has been shown to decrease with time. Thus either He^* or NF_3 must decrease to be compatible with the above reaction.

As a matter of fact, the helium metastable density rapidly reaches an equilibrium with the discharge as shown by the data in Fig. 5. Here, the optical absorption on the 3889 Å line ($2^3S_1 - 3^3P_2$) was monitored along a radial path under the same conditions as shown in Fig. 4. If anything, the metastable density is increasing with time rather than a decrease as would be required to explain Fig. 4 based upon a constant NF_3 density. Thus one is left with the alternative that the Fluorine donor is being depleted.

Laser Parameters

An important portion of this investigation was the study of the device parameters of this laser and their relationship to the discharge parameters of the hollow cathode. From the relationship between laser and discharge parameters, it is possible to infer some notion of the excitation mechanism leading to the upper state. What has been discovered is that there appears to be a different excitation process for different Fluorine donors. Details of the excitation mechanisms will be discussed in this section.

In the first portion of this section, data using SF_6 as the Fluorine donor will be discussed. This data will be related to results with NF_3; and finally, essential differences between the two will be discussed.

Figure 7 shows a plot of the laser output power as a function of the discharge current density for a 5 μs pulse. Laser power is measured with a silicon PIN-5 photodiode and represents power on all lasing lines. Current is measured using a McPhearson Rogowski coil, then divided by the surface area of the cathode to give current density, J. The data shown in Fig. 7 indicates a fourth power dependence of laser intensity upon current density. Assuming some sort of multiple step excitation mechanism in reaching the upper Fluorine excited level, the roll off or change in the functional dependence of the laser intensity with current or density indicates a saturation or limitation in one of the intermediate steps leading to the upper excited state.

A preliminary idea of the functional dependence of the laser intensity upon current may be found from looking at the following equation for an inhomogeneously (Doppler) broadened system:

$$\frac{dI_\nu}{dz} = \{\frac{\gamma(\nu)}{(1+I_\nu/I_{sat})}{}^{1/2} - \alpha\}\ I_\nu \qquad (1)$$

where I_ν is the intensity at frequency ν, $\gamma(\nu)$ is the gain coefficient, α is the loss, and I_{sat} is the saturation intensity. This equation describes the interaction of some field intensity at frequency ν with the amplifying medium provided by the discharge within the optical resonator. A more precise description of this equation will be provided later when gain parameters are discussed. It is sufficient at this time to mention only that

$$\gamma(\nu) = f(J) \qquad (2)$$

i.e., the gain coefficient is some unknown function of the current density.

Assuming that the intensity, I_ν, reaches some steady state value within the optical resonator, then:

$$I_\nu \sim \{(\frac{f(J)}{\alpha})^2 - 1\}$$

or

$$I_\nu \sim f^2(J)$$

which leads to:

$$\gamma(\nu) = f(J) \sim J^2. \qquad (3)$$

A simple experiment was performed to measure γ_o, the small signal gain at the center frequency of the transition. Microscope slides,

which act as elements of discrete loss, are inserted into the cavity between the amplifying medium and the front mirror. These slides are placed at fixed angles nearly perpendicular to the optical path and in the same plane of polarization as the Brewster windows. Each slide represents a loss of about 12% at 7039 Å. Continuous values of loss less than 12% are provided by rotating one of the slides. Small signal gain is measured as a function of current by inserting enough loss into the cavity to reach the threshold condition for oscillation. Then:

$$\gamma_o = \alpha = -\frac{1}{2\ell} \ln T_s^{\;2} R_1 R_2 \tag{4}$$

where T_s is the amount of discrete transmission provided by the slides and R_1 and R_2 are the reflectivities of the mirrors, and the cavity loss α is prorated over the length of the cathode. Figure 8 shows the results of this experiment. Plotted along with the experimental points is a line representing a square law dependence of small signal gain γ_o with current density, J. The functional dependence of γ_o to J obeys the behavior indicated by the intensity vs. current density data, namely:

$$\gamma_o \sim J^2 \tag{5}$$

with a tendency toward saturation at higher values of current density.

Consider the expression for the small signal gain coefficient

$$\gamma_o = A_{21} \frac{\lambda^2}{8\pi} g(\nu) [N_2 - \frac{g_2}{g_1} N_1] \tag{6}$$

Aside from some obvious multiplicative factors, the term $A_{21}\ N_2$ represents the spontaneous emission rate from state 2 to 1. Thus any effect due to bottlenecking in the lower state would appear as a distinct difference between the measured laser gain and the measured spontaneous emission from the upper state. However, as Fig. 8 demonstrates, the spontaneous emission exhibits the same functional dependence as does the gain.

$$N_2 \propto J^2. \tag{7}$$

Thus the pumping of the upper state appears to be a 2-step process (i.e., one requiring two electrons) which saturates at large values of current density. Furthermore, this two step excitation process, which is contrary to the previously described dissociative excitation requiring only one electron, may be a characteristic of the SF_6 donor.

Figure 9 shows a plot of laser intensity vs. current density for three different hollow cathode tubes. For this data, NF_3 is the Fluorine donor and the functional dependence of laser intensity upon current density appears to be linear as shown by the three solid lines. This behavior is radically different than that observed for SF_6 and indicates the possibility of two different excitation mechanisms for the two donors.

One Step and Two Step Excitation

Several authors[8,11,13] have suggested that dissociative excitation pumps the upper states of Fluorine:

$$He(2^3S) + SF_6 \rightarrow F^{**}(3p\,^2P) + He(1s) + SF_5^{*}. \tag{8}$$

If the Helium metastable is assumed to be pumped by direct electron impact in the negative glow:

$$e^- + He(1s) \Rightarrow He(2^3S) + e \qquad (9)$$

then pumping of the upper state of Fluorine is a "single step" process (where single indicates dependence upon a single electron).

An alternative to the dissociative excitation process is ionic recombination:

$$He^+ + \left\{ {SF_6^- \atop SF_5^-} \right\} \rightarrow (HeF)^* \rightarrow F^{**}\ (3p\ ^2P) + He + SF_5^* \qquad (10)$$

This type of reaction would require two electrons (2 step process); one to produce He^+ and another to be attached to the Fluorine donor.

Two body, ion-ion recombination (or charge neutralization:

$$A^+ + B^- \rightarrow A^* + B \qquad (11)$$

has been reported by several authors[20-25] as having a very large cross section ($\sim 10^{-13} - 10^{-14}$ cm^2) at low energies ($<$ 2 eV). Pororelli and Shukhtin[24] reported that the hollow cathode discharge was an ideal medium for ion-ion recombination. Miller and Morgner[25] predict large cross sections for the reaction:

$$He^+ + F^- \rightarrow He + F^* \qquad (12)$$

producing various excited states in Fluorine.

The ion-ion interaction:

$$He^+ + SF_6^- \twoheadrightarrow F^{**}(3p\,^2P) + He + SF_5 \qquad (13)$$

appears plausible for two reasons: (1) the bulk of the energy for this reaction is carried away by the excited Fluorine atom, moreover, any additional energy can be accounted for by the remaining two species (primarily SF_5^* which has a myriad of internal modes to absorb energy); and (2) conditions in the hollow cathode favor the production of the two ionic species [He^+] and [SF_6^-], since there is an abundance of energetic electrons to produce the former and many secondaries to produce the latter. Additional evidence for the first was implied in the results obtained by Hocker[32] in his high spectral resolution study of the Fluorine laser. He found an effective temperature of 2200 - 3200° K to describe the Doppler width of the upper state of the 7037 Å transition.

The hollow cathode discharge, due to its non-Maxwellian electron energy distribution, is a very favorable medium for the production of both the ionic species in Eq. 20. Belal and Dunn,[26] using a laser heterodyne technique, measured electron and ion densities as high as 10^{14} cm^{-3} in a hollow cathode discharge in He. Furthermore, in a similar He hollow cathode discharge, McIntosh, et al.[27] measured the He (2^3S) densities to be less than 10^{12} cm^{-3}. This indicates that the ratio of He^+/He (2^3S) > 10^2, which, incidentally, is why the Duffenback reaction: $He^+ + Cd \rightarrow He + (Cd^+)^*$ dominates over Penning ionization in the hollow cathode, He - Cd^+ laser.

In the case of SF_6, much work has been done in studying the negative ions formed from this compound. In particular, the following two electron attachment processes are thought to occur at low electron energies (< 1.5 eV):[28-31]

$$SF_6 + e \rightarrow \begin{cases} SF_6^- \\ SF_5^- + F \end{cases} \quad . \tag{14}$$

Furthermore, in the case of the electron resonant capture formation of SF_6^-, the cross section[28] is reported to be > 10^{15} cm^2 at electron energies of 0.1 eV. Therefore, with the large production rate of secondary electrons, the rate for the negative ion formation reaction in Eq. 14 should also be very large.

The effects of this comparatively high attachment rate in SF_6 is shown in Fig. 10. In the first photograph, spontaneous afterglow light on the 3889 Å transition of He is shown for a mixture of He + NF_3. It can be seen by the afterglow light there is strong indication of electron-ion recombination even in the presence of NF_3. However, with an equal amount of SF_6 substituted for NF_3, the afterglow light is completely quenched indicating the absence of substantial e-i recombination events in the afterglow.

In view of the fact that Eq. 13 predicts the correct functional dependence of gain (and spontaneous emission), the ionic channel appears to be the primary route for the excitation of the upper state in He-SF_6 mixtures. It is also noted that:

(1) from the standpoint of conservation of energy the two reactions are comparatively equal;

(2) the large cross sections for binary ion-ion recombination reactions in other species favor this mechanism;

(3) the densities of the reacting species favor the ionic mechanism, i.e., $[SF_6^-]\,[He^+] \gg [SF_6]\,[He^*]$ in a hollow cathode;

(4) there is some experimental evidence supporting a two-step pumping mechanism for the upper laser state.

As was mentioned earlier, the results found here for SF_6 and the conclusions based upon these results are not meant to be extrapolated to other Fluorine donors. For example, the data shown in Fig. 9 for NF_3 does not indicate the same 2-step process as was assigned to SF_6. The fact that SF_6 appears to be a much poorer donor than NF_3 is reflected in the lower intensities obtained with SF_6 (about an order of magnitude lower). Figure 11 shows photographs comparing the formation time of the population inversion. In the case of NF_3, lasing occurs at about 1.5 μs from the beginning of the current pulse. However, for the case of SF_6 lasing begins at about 4 μs for identical He and Fluorine donor gas mixtures. However, this difference is somewhat obscured by the performance of the discharge. Inasmuch as the energy reaches the negative glow portion via the energetic beam electrons accelerated by the cathode fall, the hollow cathode discharge expressed a definite preference for the gas with the lower mass density.

V. Summary and Concluding Remarks

The hollow cathode discharge has proven to be a very useful device for exciting gas mixtures containing highly electronegative species. With such a device it has been possible to couple large amounts

of power (>/kW/cm^3) into mixtures containing SF_6, NF_3, CCl_4, CCl_2F_2, I_2, or virtually any halide compound and obtain stable, repetitive pulses of greater than 100 μs duration. This ability to maintain very uniform excitation in such strongly electron attaching gases is due to the beam nature of the excitation in the negative glow region which is the majority of the hollow cathode discharge.

Employing these useful hollow cathode properties, lasing was obtained in mixtures of He and a Fluorine donor compound (SF_6 or NF_3). Lasing was obtained on six lines in the near infrared with some new results, previously unreported in positive column operated lasers. Whereas operation of this laser in a positive column device has been limited to a very short ($\leq$ 2μsec), pulsed mode. Lasing in the hollow cathode configuration has been accomplished for pulse durations on the order of 100 μs. This suggests that CW operation could be obtained in a transverse flow configuration.

There are several possible explanations for the difference in pulse length capabilities between the two types of tubes. Resonant trapping by the lower state has been mentioned as the limiting factor by previous authors. This would occur as a result of a substantial amount of free Fluorine to trap the resonant radiation emitted by the lower laser state. As was discussed in Section VI, it is likely that there are more free Fluorine atoms available in a positive column discharge than in the negative glow due to the nature of the electron energy distribution.

Another possible explanation for the termination of the laser pulse in the conventional configuration is that a positive column discharge tends to constrict when operated with electronegative gases. This tendency toward constriction is due to the presence of strongly electron attaching species, which may precipitate instabilities in the

discharge. However, in the hollow cathode configuration, excitation occurs by beam electrons so that there is no electric field dependent equilibrium to be affected by instabilities.

A third explanation may be that the Fluorine donor is depleted much more rapidly in the positive column discharge. This rapid depletion would be the result of the higher current density in the small bore, positive column tube. In the case of the hollow cathode, the only obstacle in the way of CW operation of this laser appears to be depletion of the Fluorine donor. The solution to this problem, then, would be to design and operate a hollow cathode, Fluorine laser in a fast, transverse flowing configuration. In this manner, the Fluorine donor could be replenished at a rate that compensates for the depletion which occurs in the excitation process. A properly designed system employing an optimized optical cavity and the most efficient Fluorine donor (HF, F_2, or NF_3) could conceivably produce tens of watts of CW power in the near infrared at an efficiency of $\sim$ 0.1%.

In addition to the long pulse operation of this laser, some new information on the excitation mechanism for populating the upper laser state has been inferred. In the positive column discharge dissociative excitation was thought to be responsible for pumping the upper laser state. Data obtained from this hollow cathode laser in mixtures containing SF_6 indicate that a bimolecular, charge neutralization reaction may be the important pumping mechanism, at least in the case of SF_6.

Although the hollow cathode discharge has been used for many years for the charge transfer type of metal vapor ion laser, many of its characteristics are naturally mated to the study of the more difficult gases such as used here.

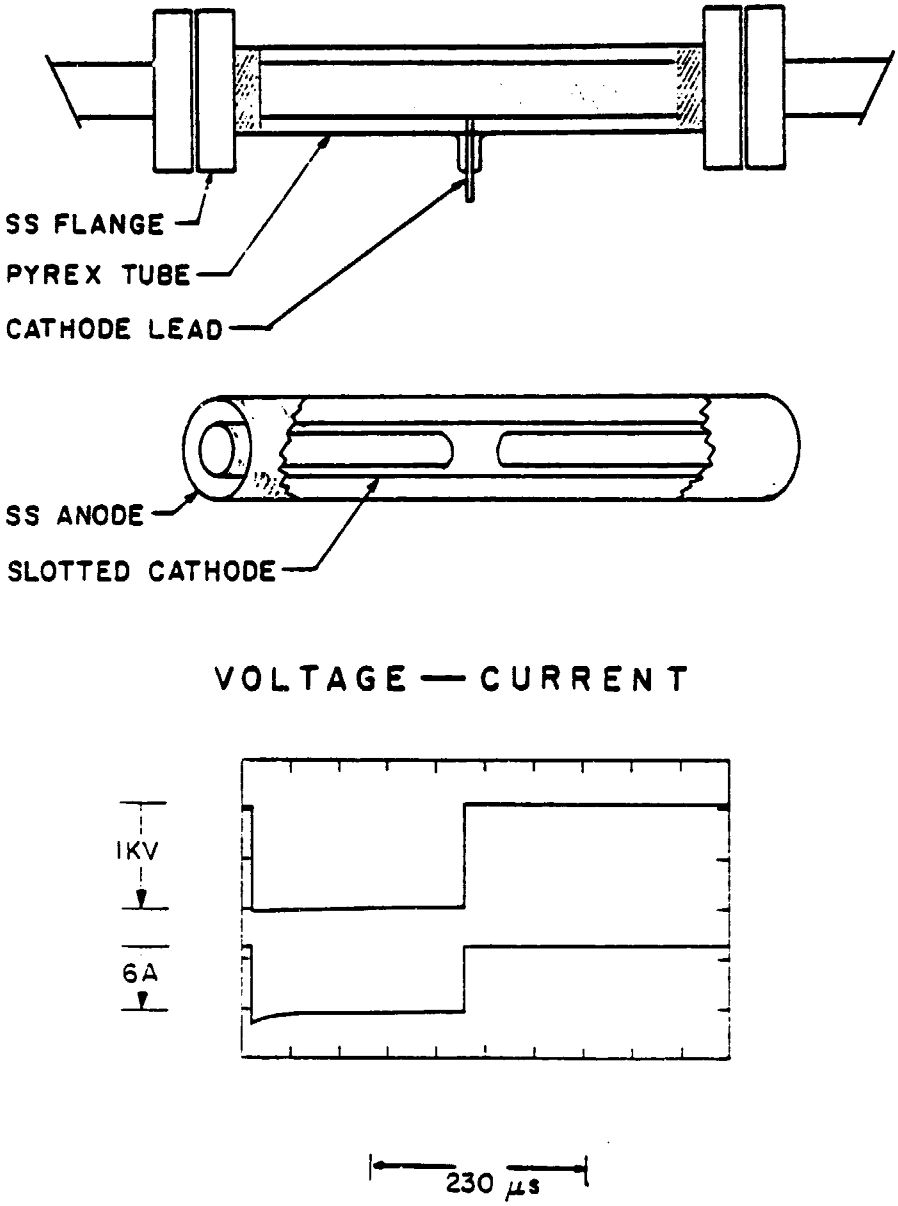

Fig. 1. Hollow Cathode discharge in pure SF_6.

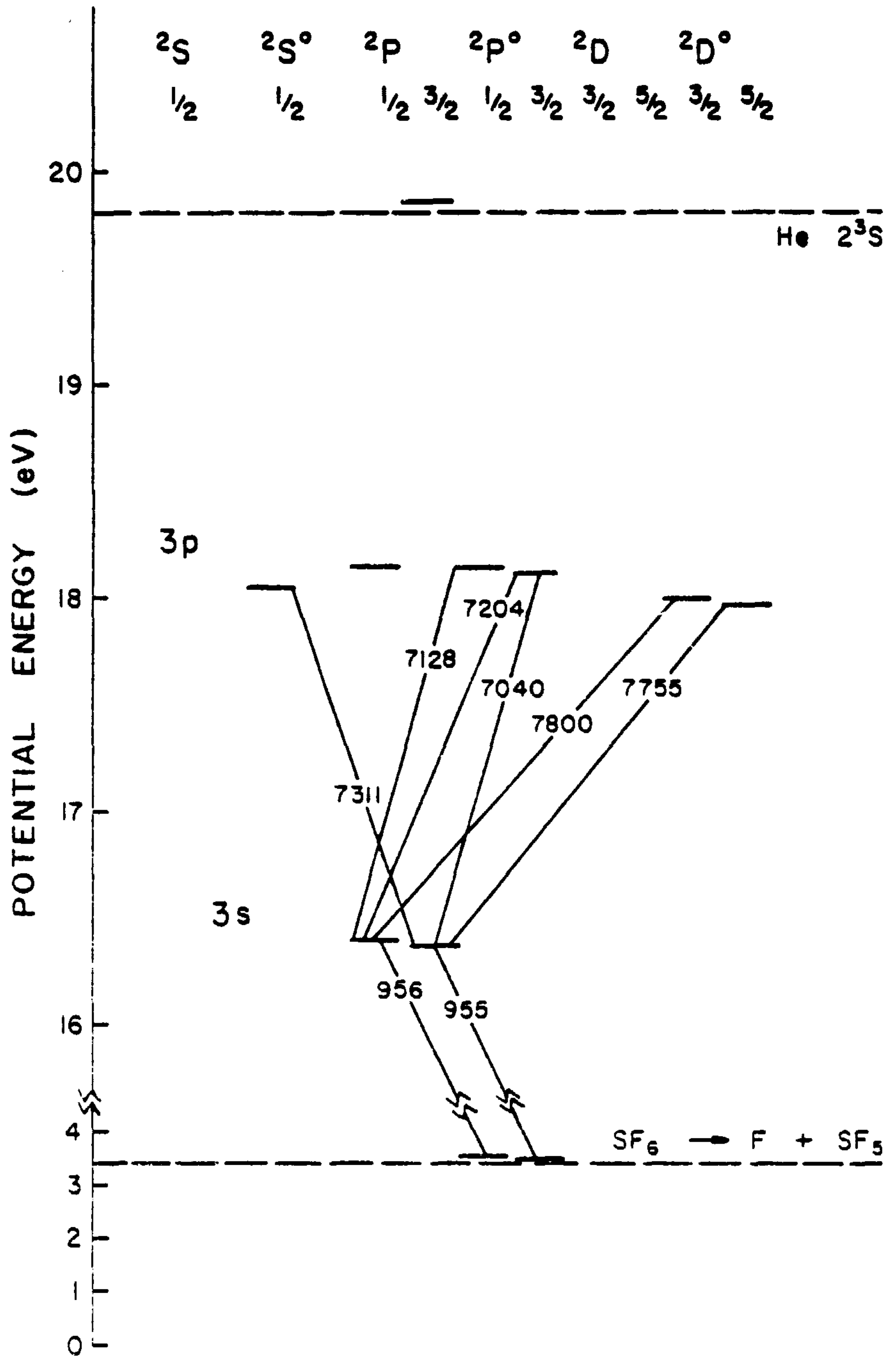

Fig. 2. Abbreviated energy level diagram for doublet system in Fluorine shown relative to dissociation energy of SF_6.

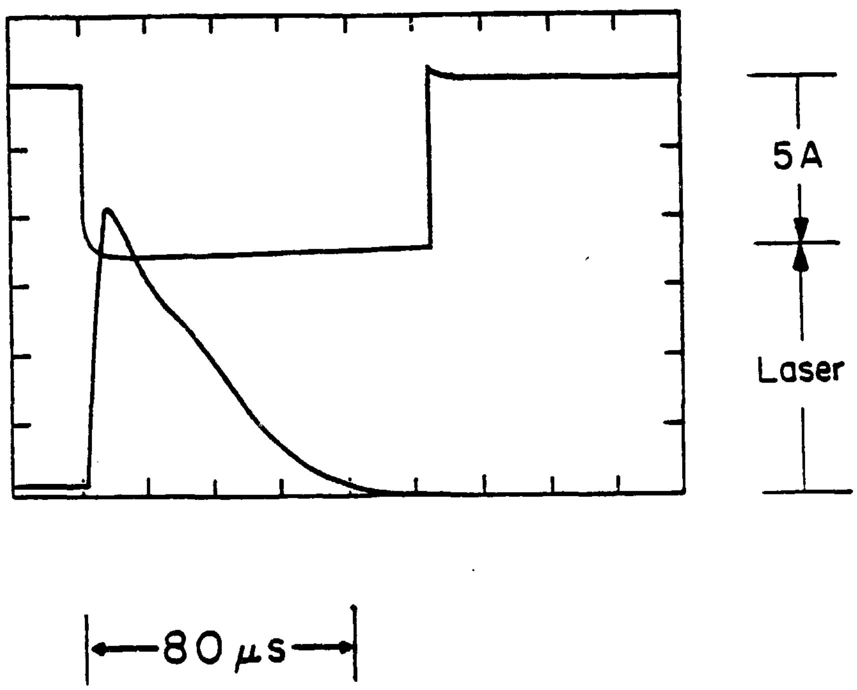

Fig. 3. The photograph shows a laser pulse lasting for 80 μsec. These data were taken with 7.5 Torr of Helium and 100 m Torr of NF_3. 1.7 cm x 100 cm cathode.

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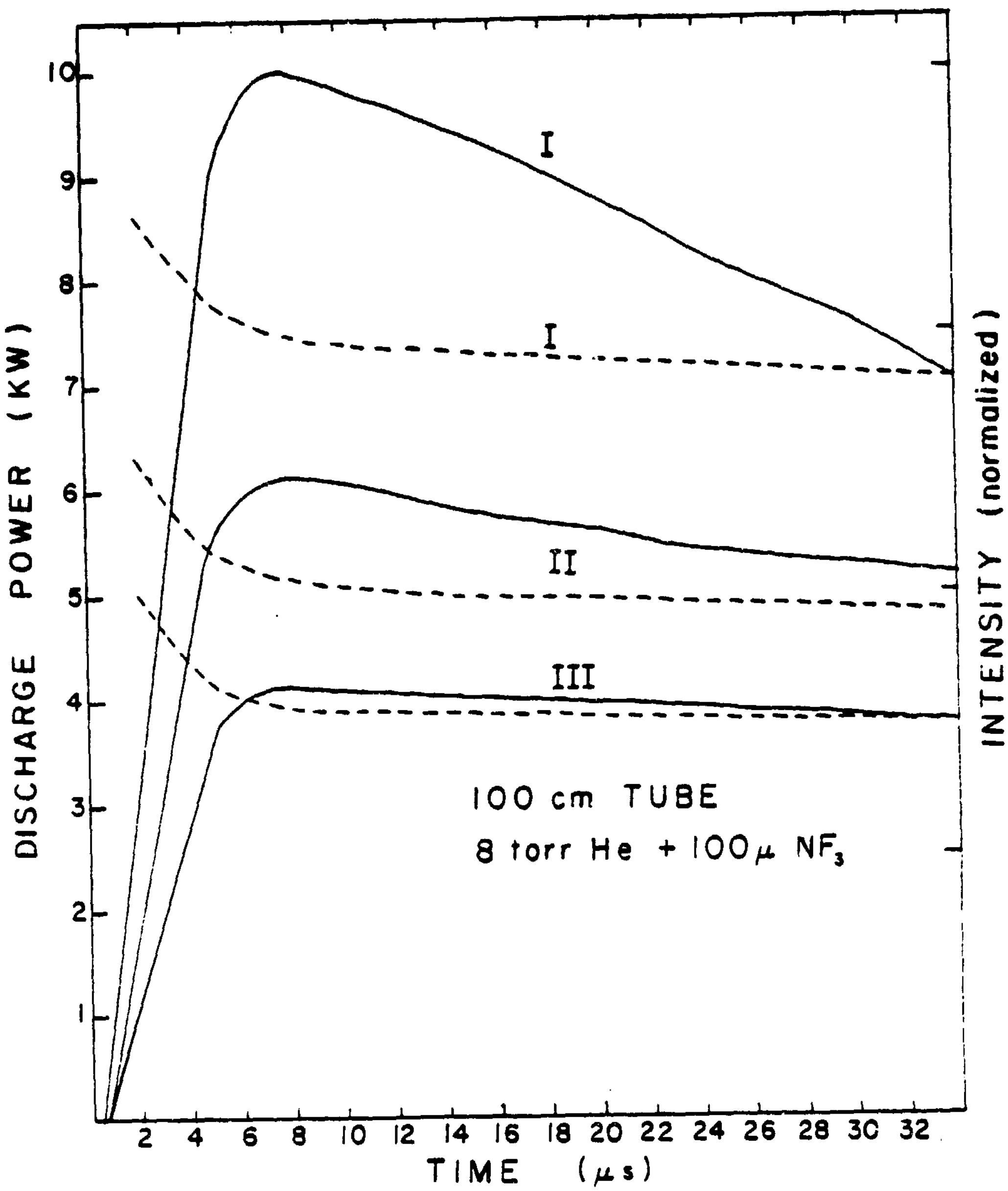

Fig. 4. This graph shows the rate of decay of spontaneous emission, 7129 A, for different values of energy into the discharge. The dotted line is discharge power; solid line is spontaneous emission.

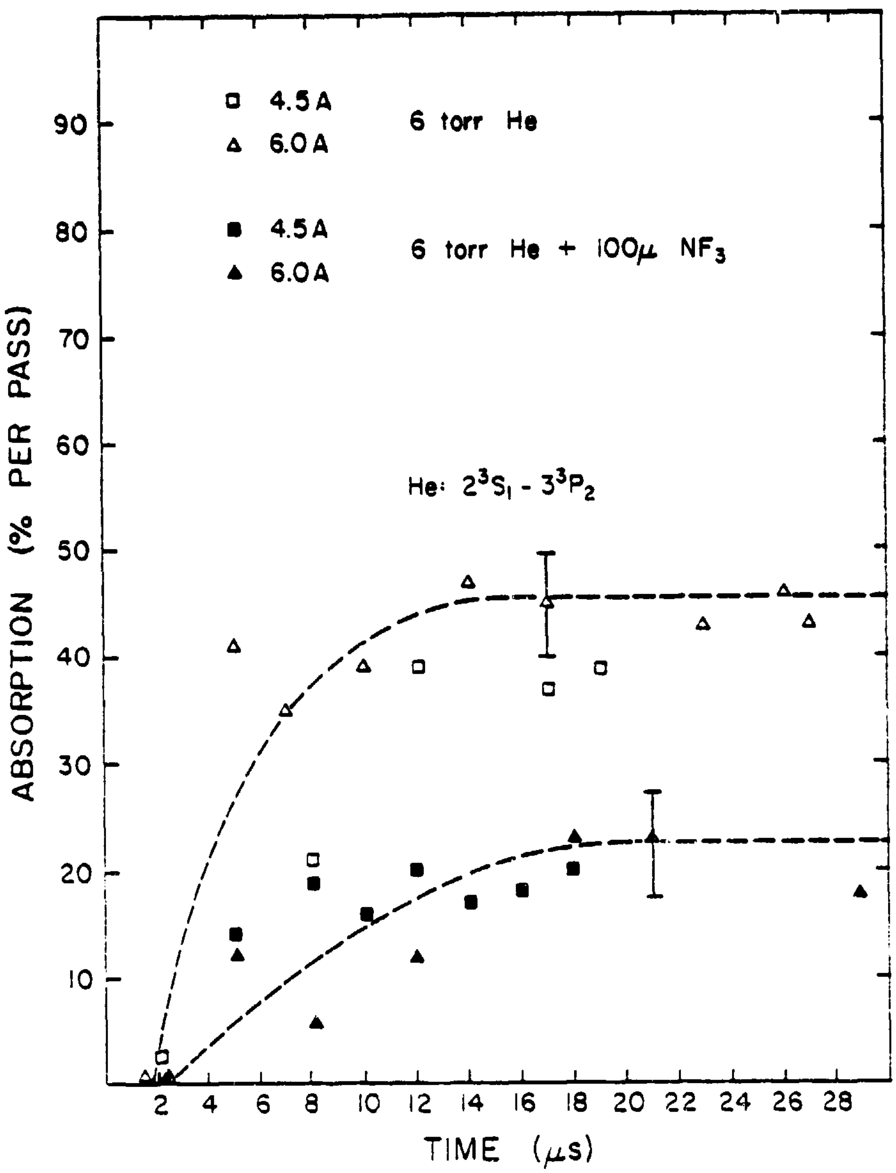

Fig. 5. The time evolutuion of the density of helium metastables in a pulsed discharge.

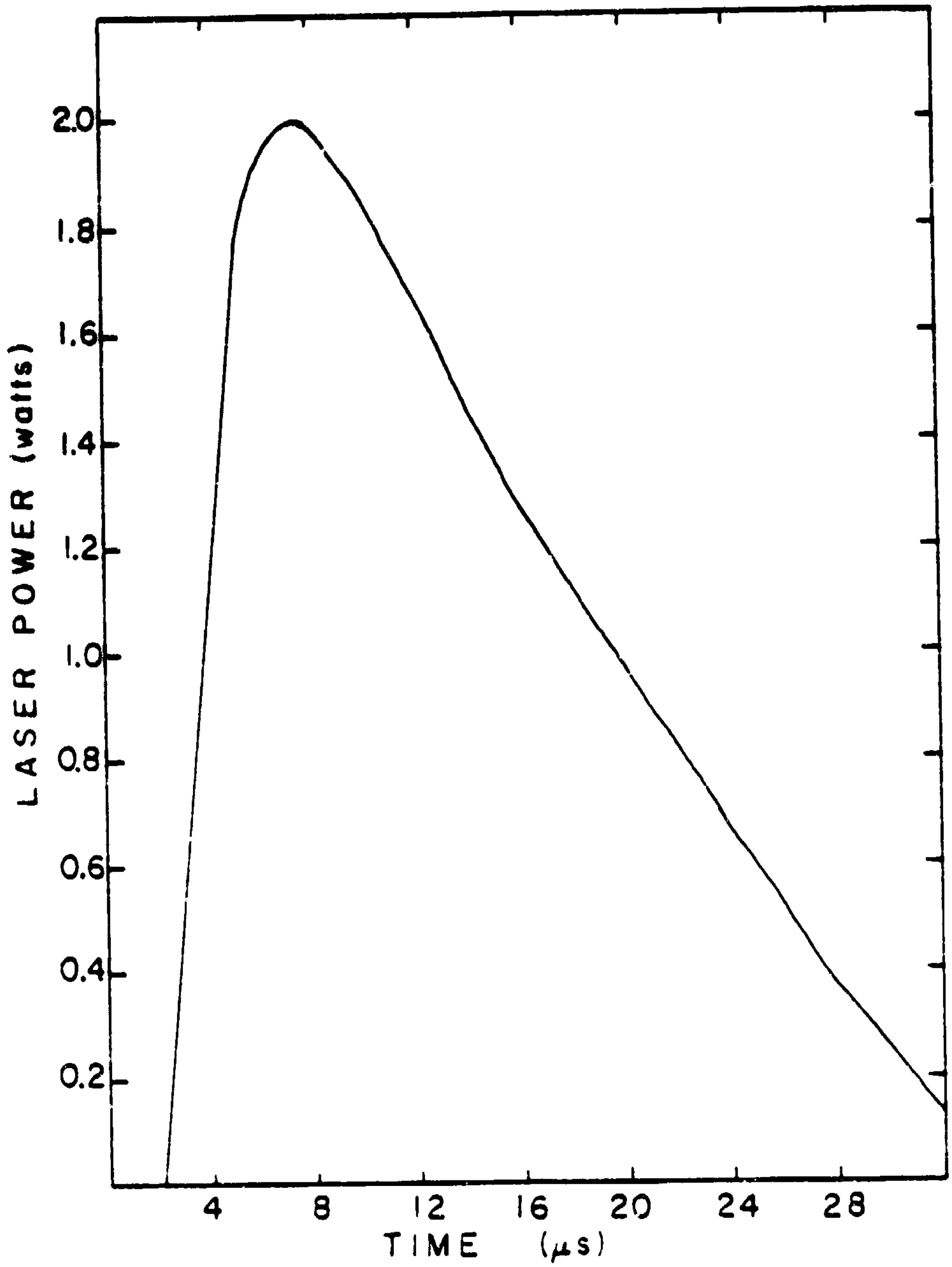

Fig. 6. The build-up and decay of the laser pulse generated by the discharge used to obtain the data in Figs. 4 and 5.

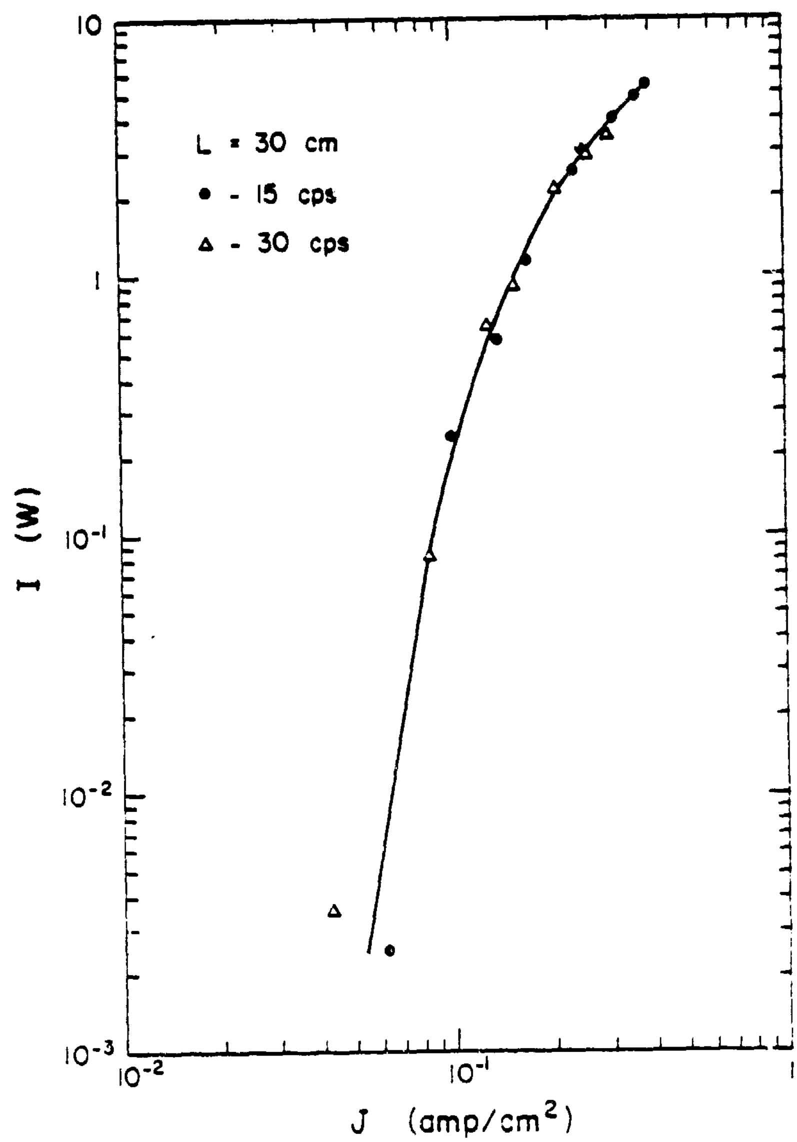

Fig. 7. Laser intensity (all lines) as a function of discharge current density for 3 Torr He + 100 μ SF_6.

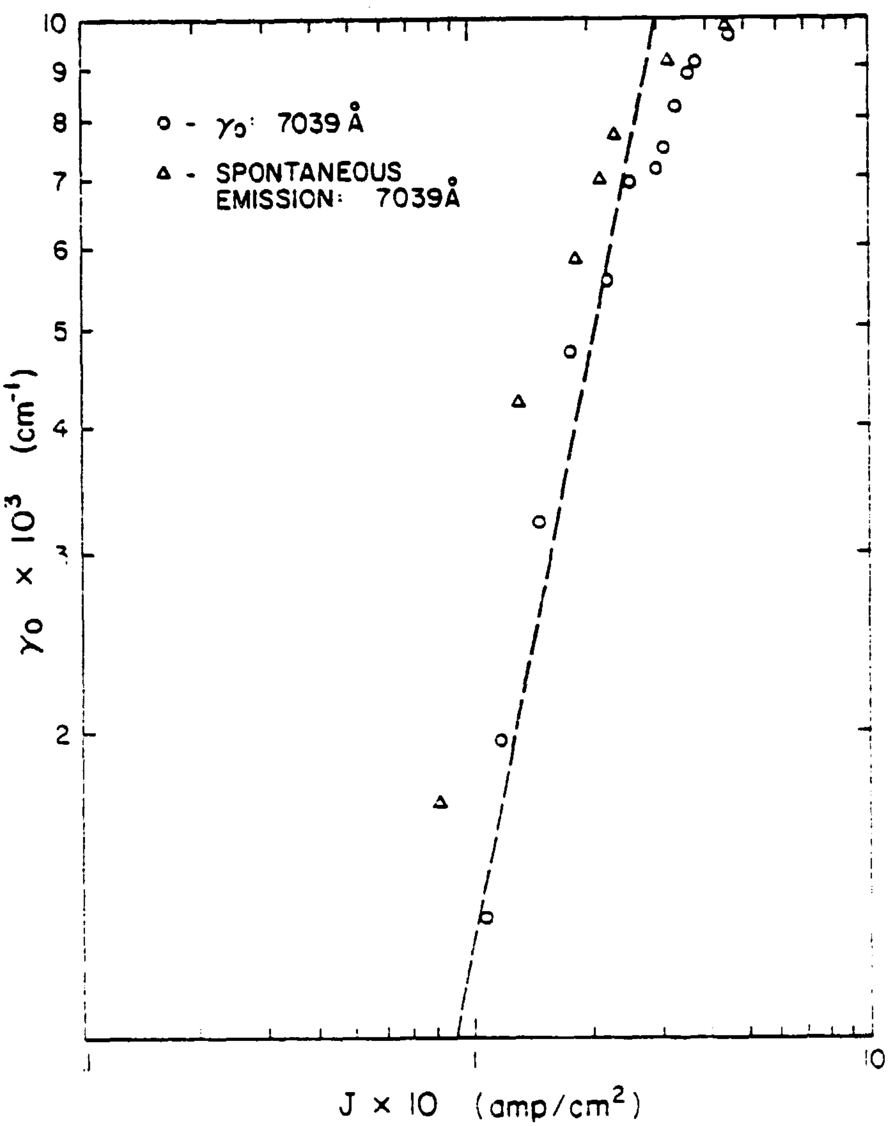

Fig. 8. Threshold gain and spontaneous emission vs. current density.

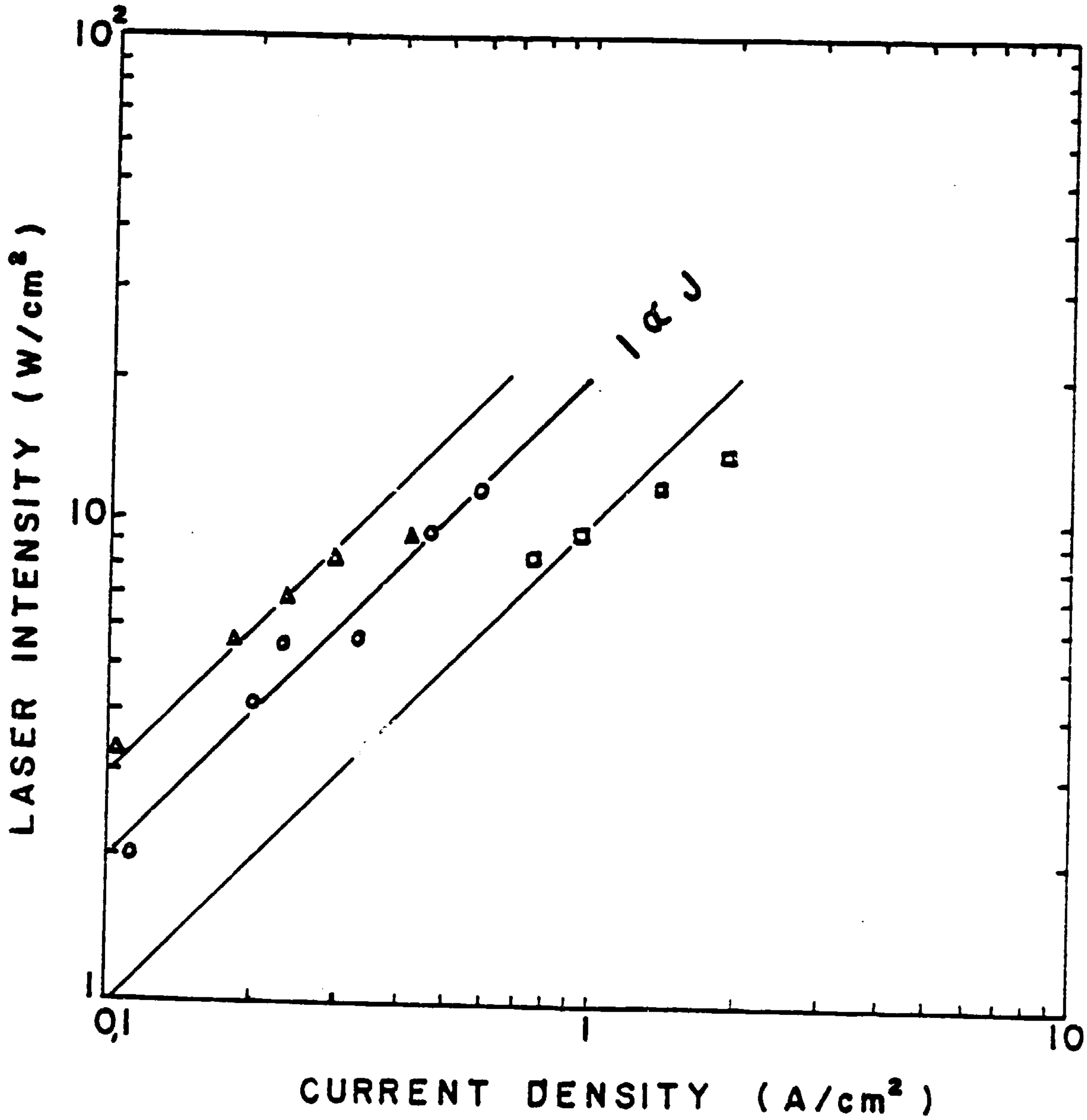

Fig. 9. Laser intensity vs. current density for three different tubes. NF_3 is the donor.

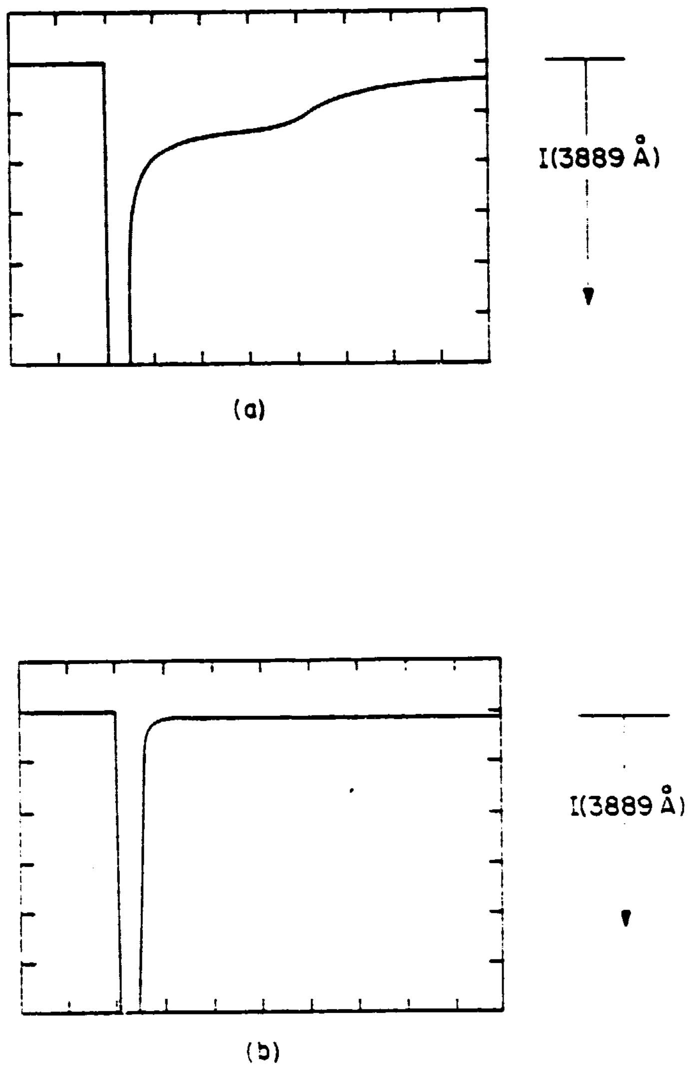

Fig. 10. The tracings show the behavior of the 3889 Å lines of helium in the afterglow of a pulsed discharge. In both cases, the helium pressure was 7.0 Torr; in (a) the donor was NF_3 at 100 m Torr and (b) used SF_6 at 100 m Torr.

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